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Abstract

Sample preparation is always a critical step in study of micrometer sized astromaterials available for study in the laboratory, whether their subsequent analysis is by electron microscopy or secondary ion mass spectrometry. A focused beam of gallium ions has been used to prepare electron transparent sections from an interplanetary dust particle, as part of an integrated analysis protocol to maximize the mineralogical, elemental, isotopic and spectroscopic information extracted from one individual particle. In addition, focused ion beam techniques have been employed to extract cometary residue preserved on the rims and walls of micro-craters in 1100 series aluminum foils that were wrapped around the sample tray assembly on the Stardust cometary sample collector. Non-ideal surface geometries and inconveniently located regions of interest required creative solutions. These include support pillar construction and relocation of a significant portion of sample to access a region of interest. Serial sectioning, in a manner similar to ultramicrotomy, is a significant development and further demonstrates the unique capabilities of focused ion beam microscopy for sample preparation of astromaterials.

Introduction

Sample preparation of meteoritic material, particularly micrometer-sized interplanetary dust particles (IDPs) or the cometary dust particles recently returned by NASA's Stardust mission (Brownlee et al., 2006), for detailed micro-analysis using electron microscopy, secondary ion mass spectrometry (SIMS) or synchrotron-based techniques (e.g., infrared spectroscopy) has long been a fundamental challenge. A variety of novel techniques have been developed and applied to this problem over the past three decades. For IDPs, a significant development was the ability to produce electron transparent (50-100 nm thick) serial sections of individual particles using ultramicrotomy techniques (e.g., Bradley and Brownlee, 1986; Bradley, 1988). Ultramicrotomy involves slicing an embedded sample with tens-of-nanometers precision using a diamond blade. Since its first utilization in astromaterials research, ultramicrotomy has been the "method of choice" for IDP sample preparation whenever such samples were intended for transmission electron microscopy (TEM) studies, although the technique has been modified over the years to enable new analytical measurements. For example, use of sulfur as an embedding medium rather than traditional low viscosity epoxy alleviated the carbon contamination problem in the preparation medium supporting IDP sections, thereby enabling measurements of the indigenous carbon content (e.g., Bradley et al., 1993). In recent years, other modifications of the embedding medium have included the use of acrylic polymers (Matrajt and Brownlee, 2006), yet the principal mechanism for sectioning, i.e. ultramicrotomy, has remained the same (e.g., Joswiak and Brownlee, 2006).

Since the late 1990s, focused ion beam (FIB) microscopy has been utilized in both sample preparation and analysis across a diverse range of scientific fields including

meteoritics (e.g., Heaney et al., 2001). A detailed description of FIB instrumentation is given in Young and Moore (2005). The technique uses a focused beam of gallium ions to ablate volumes of material at controlled rates and at precise locations within a sample. This FIB milling can be used to prepare electron transparent sections that can be removed from the bulk sample using either an external glass needle micromanipulator under an optical microscope (e.g., Lee et al., 2003) or a manipulator device within the instrument chamber (e.g. Graham et al., 2006; Zega and Stroud, 2006). The sections are then subsequently mounted on TEM grids. It is the ability of the FIB technique to prepare site-specific sections from a sample that has had the greatest impact on meteoritic sample preparation. Stroud et al. (2004) highlighted integrated studies of presolar grains from meteorites initially using SIMS for isotopic characterization, followed by use of FIB to extract a cross-sectional slice containing the isotopically anomalous material. The section may subsequently be studied using TEM for structural and elemental observations. This integrated approach proved to be a significant advance as previous studies required complex and time-consuming ultramicrotomy to produce planar TEM sections from an entire sample that had initially been characterized using SIMS (Keller et al., 2004). Herein we discuss the application of FIB to support: (i) integrated studies of an IDP using NanoSIMS, TEM and synchrotron FTIR analytical techniques and (ii) extraction of cometary residue material deposited within micro-craters by hypervelocity capture during the Stardust mission.

Methods and Materials

The IDP L2047,D23 was provided by the cosmic dust curatorial facility at the NASA

Johnson Space Center; details of the stratospheric collection, curation and initial characterization of IDPs are given by Warren and Zolensky (1994). Prior to isotopic imaging at Lawrence Livermore National Laboratory using a Cameca NanoSIMS 50 ion microprobe, L2047,D23 was washed in hexane to remove remaining silicone collector oil. Silicon micro-tweezers were used to remove the particle from the dimpled glass slide in which it was transported from the cosmic dust curatorial facility, and it was then transferred to high-purity Au foil. The particle was pressed into the foil to produce a flat surface suitable for isotopic imaging. Details of the isotopic imaging protocol using NanoSIMS are given in Bradley et al. (2005) and Smith et al. (2005). The FIB extraction of material from L2047,D23 was carried out at FEI Company's Hillsboro demonstration facility using an FEI Strata^{TM} DB-STEM 237 focused ion beam/field-emission scanning electron microscope fitted with an EDAX Genesis energy-dispersive spectrometer (EDS), Omniprobe^{TM} in-situ micromanipulator extraction system and a four-region (bright field, dark field and two high-angle dark field) SEM-STEM detector. The FIB-extracted electron transparent section of L2047,D23 was analyzed using a Philips CM300 300 kV FEG-TEM fitted with an Oxford Inca EDS. The Fourier-Transform Infrared (FTIR) spectroscopic measurements were collected using IR microspectroscopy beamline 1.4.3 at the Advanced Light Source, Lawrence Berkeley National Laboratory.

Aluminum foils containing the micro-craters generated by cometary dust particle impacts at 6 km/s were recovered from the Stardust sample tray assembly (Hörz et al,. 2006). The two foils (C2086W,1 and C2091N,1) were initially characterized at the Natural History Museum (UK) using a JEOL 5900LV scanning electron microscope (SEM) fitted with an Oxford Inca EDS. The typical analysis conditions were 20 kV and

2 nA. The Cameca NanoSIMS 50 ion microprobe at Washington University in St Louis was used to determine the C, N, and O isotopic composition of the cometary material at high spatial resolution on the rim of a ~240 μm diameter impact crater on the surface of foil C2086W,1. Full details of the analytical protocols are given in McKeegan et al. (2006) and Stadermann et al. (2007). FIB extraction of cometary residue material from the two foils was performed at Lawrence Livermore National Laboratory using an FEI Nova 600 Nanolab dualbeam FIB/FESEM microscope with an EDAX Genesis EDS and both an Omniprobe™ in-situ micromanipulator and an Ascend Instruments Extreme Access™ extraction system.

Integrated Studies of an IDP

IDPs represent some of the most primitive and small-scale extraterrestrial material available to study in the laboratory (Bradley, 2005). To fully understand an individual IDP, it is necessary to study the different mineralogical, chemical and isotopic properties which make up the overall composition. As has been shown previously combined isotopic and mineralogical studies of meteorites and IDPs can yield new insights into the processes of their formation (Stroud et al., 2004; Keller et al., 2004; Bradley et al., 2005). The isotopic images acquired from the analysis of the surface of L2047,D23 using the NanoSIMS identified a small, ~800 nm diameter grain with a large ^{15}N enrichment, $^{14}N/^{15}N$ ratio of 194±4 compared to the (normal) ratio acquired for the whole particle of 272±2. The combination of the NanoSIMS measurements and subsequent examination of the IDP using FESEM identified the location of the anomalous grain (Fig. 1a). To

understand the anomalous grain in the context of its own composition and that of the surrounding material, a FIB-TEM section from the area was prepared.

In addition to the ability to mill or ablate material away, the interaction of ions (and indeed electrons) with introduced gases of specific compositions can result in deposition of material onto the surface of the sample (Stevie et al., 2005). In this study, this capability was used to deposit a protective layer of Pt over the well-defined region of interest (ROI) containing the grain. These protective layers of so-called "strap" assist in reducing ion beam damage to the ROI during FIB milling. In-depth discussion of the FIB milling process in preparation of electron transparent sections from bulk materials can be found in papers by Heaney et al. (2001); Lee et al. (2003); and Anderson and Klepeis (2004). For our work, after deposition of the protective Pt "strap", a 30 kV focused Ga ion beam (approximately 5000 pA current) was used to mill trenches in the material on either side of the strap, leaving a section ~1 μm thick (Fig. 1b). The beam current was then reduced to 300 pA to make side-wall cuts and an undercut through the thinned section, to release it and enable extraction with the in-situ micromanipulator needle (Fig. 1c). Once extracted from the bulk sample, the section was moved to a specially constructed semi-circular Cu TEM half-grid, also in the chamber. The section was attached to the TEM grid by depositing Pt to form a so-called "weld", and the FIB was then used to cut the micromanipulator tip away from the section, leaving the section attached to the grid. The section was then thinned using the 30 kV FIB at 300 pA beam current; once "electron transparency" was approached the current was reduced to 100 pA to minimize surface damage in the finished section. Thinning resulted in a section approximately 80-100 nm thick (Fig 1d).

As with ultramicrotomy, FIB preparation of an electron transparent section can result in a number of artifacts in the sample; these are discussed in both Lee et al. (2003) and Anderson and Klepeis (2004). For astromaterials, there are several significant issues related to the production of thin sections using FIB. (i) Ga+ ions are implanted into the sample during the milling process. This is problematic for detailed X-ray microanalysis by EDS, as the Ga L-lines lie very close to the mineralogically important Mg K-lines. However, modern spectral processing software is often capable of extracting or removing unwanted artifact peaks which are not indigenous to the sample. (ii) Potential specimen damage during the FIB milling and polishing stages can form amorphous layer a few nanometers (less than 10 nm) thick. This problem can be mitigated by the use of protective Pt and C "straps" deposited prior to milling, and by performing a low voltage (5 kV) polishing at the end of the sample preparation (Giannuzzi, 2006). (iii) There is the potential for loss or damage of volatile material such as organic molecules. However, a number of recent papers studying biological material have included structural studies on samples prepared using FIB and have found little evidence of significant FIB-induced damage of organics (Heymann et al., 2006). (iv) FIB requires specialized skills and expensive instrumentation compared to sample preparation techniques such as ultramicrotomy. (v) Depending on the composition of the sample and the nature of the lift-out, the FIB extraction process can be more time consuming than other techniques, although it is unlikely that these could offer comparable accuracy in site-specific extraction of samples. (vi) Material surrounding the ROI is consumed during the trenching stages of the FIB milling making it more difficult to extract serial sections. Solutions to this latter problem are discussed below.

An electron transparent section can be characterized using a number of analytical techniques including TEM, FESEM and NanoSIMS. In the example discussed here, the section was initially imaged using the 30 kV SEM-STEM detector permanently mounted in the dual-beam microscope chamber. The use of SEM-STEM in astromaterials research is still in its infancy, although recent studies have shown the potential of the technique (Lee and Smith, 2006). While the SEM-STEM detector does not have the resolution of a conventional TEM, the images acquired in real time allow a monitoring of specimen thickness during the final stages of thinning, i.e., if the material within the section can be imaged at 30 kV, then the section is sufficiently thin to be imaged in the TEM at 200-300 kV. For example, in the SEM-STEM bright-field image of the IDP section, it was possible to: i) see a small grain at the top of the section that could be the isotopically anomalous feature and ii) texturally infer that the section is dominated by a layer silicate phase (Fig. 1e). The subsequent detailed TEM analysis of the section confirmed the layer silicate phase to be serpentine, based on the ~0.7 nm basal lattice fringe spacing acquired using HRTEM. The small grain in the top surface of the section was identified as amorphous carbon. In addition to the detailed TEM study and re-examination using the NanoSIMS, the FIB section can also be used for a number of other analytical techniques, such as synchrotron X-ray fluorescence and IR spectroscopy. In this example, prior to destructive re-examination using NanoSIMS, the section was subject to infrared spectroscopy to further characterize the amorphous carbon grain (Bradley et al., 2005), and which also identified the serpentine phase (Fig. 1e). Further analysis of the section in the NanoSIMS confirmed that the amorphous carbon grain did contain the ^{15}N enrichment (Fig. 2). The implications of the ^{15}N enrichment are

discussed in detail in Smith et al. (2005) and the overall significance and context of this integrated study are discussed in Bradley et al. (2005). This example shows it is possible to derive mineralogical, isotopic and optical spectroscopic information, the latter enabling direct comparisons with astronomical observations, all from a single FIB-TEM section. The ability to perform integrated studies on the nano-scale phases within individual particles will significantly impact future astromaterials research, for example, with sample return missions such as Stardust where there are only nano-grams of material in each sample available to study.

Extracting Cometary Residue from Micro-craters

The hypervelocity capture of Wild-2 cometary dust (at ~6.1 km/s) in the low-density silica aerogel and on the additional surface provided by the aluminum 1100 series foil wrapped around the sample tray assembly presented new challenges for sample preparation to enable detailed analytical measurements of these precious particles. From the aerogel, individual impact tracks were extracted from the bulk tiles using micro-needles (Westphal et al., 2004) and ultra-sonic blades (Ishii et al., 2005; Ishii and Bradley, 2006). Once extracted, ultramicrotomy can be used to prepare thin-sections suitable for analysis by multiple techniques, e.g., TEM, FTIR and NanoSIMS (Matrajt and Brownlee, 2006). For craters preserved on aluminum, techniques for extraction of residue material remaining from the original impactor were developed during LDEF (Long Duration Exposure Facility) studies by Teetsov and Bradley (1986), who used a combination of micro-replication and ultramicrotomy. More recent efforts by Leroux et al. (2006) and Graham et al. (2006) have shown that FIB techniques can be used to extract complete

cross-sectional slices of craters (less than ~10 μm diameter) or recover individual residue fragments and melt droplets. In this paper we describe expansion of, and modification to, the recovery technique discussed in Graham et al. (2006), as applied to "real" Stardust craters, not laboratory analogues.

The optical and electron microscopy studies of the aluminum foil during the preliminary examination period (January to August 2006) identified a diverse range of craters from several hundreds of micrometers to sub-micrometer in diameter (Hörz et al., 2006). During the Stardust preliminary examination, only a limited number of craters greater than 50 μm diameter were released for study (Hörz et al., 2006). To maximize the available compositional information from the preserved cometary residues, these craters were analyzed as a part of a multi-technique consortium that included isotopic measurements.

Extraction of Residue Material from Crater Rims

The preliminary SEM-EDS analysis of the crater from foil C2091N,1 identified abundant residue material (Fig 3a). However, residue material was deposited on the base, side-walls and rim/lip of the crater, across very complex topography that did not allow the use of the "classical" lift-out methodology discussed in the previous section. Also, it was highly desirable not to consume substantial amounts of the precious Stardust residue during FIB milling to generate a section, or to deposit reworked material elsewhere in the crater. An ROI (region of interest) containing Mg-rich residue was found on the lip of the crater (Fig 3a). Prior to the FIB milling, both C- and Pt-straps were deposited over the ROI. A C-strap was deposited first, to physically isolate the residue from overlying Pt,

and thereby reduce possible interfering EDS peak overlap between Pt M-lines and the K-lines from any sulfur that might be in the cometary dust residue. The selection of a ROI located on the crater lip (rather than on the crater wall or base) took advantage of the raised position that yielded a relatively simple geometry for section retrieval, and a free rear surface. In the "classical" FIB-TEM preparation method, trenches are made on either side of the section to enable side-wall cuts and, more importantly, to permit an oblique angle undercut that releases the section from the bulk material. Although highly variable and sample/material dependant, for a lift-out section which is 20 μm in length, the trenched boxes either side of the section typically are 20 μm in length by 10 μm in width, and are milled to a depth of approximately 7 μm. The FIB trenching step in the preparation sequence typically results in the destruction of a substantial amount of the sample, particularly if the ROI is only a few micrometers in diameter. However, in this Stardust foil example the position of the raised and overturned crater lip, above an undisturbed foil surface, meant that the section required no undercut to break the section free from the substrate. As a result, it was only necessary to mill initial, narrow, vertical 30 kV FIB cuts at 1000 pA beam to cut through the entire depth of the overturned crater lip on either side of the ROI (Fig. 3b). This slicing method of sectioning provides an important advantage because it dramatically reduces the area of damage and volume of material lost during the initial preparation stage. However, with no underlying substrate supporting the section, there was a high risk of section collapse and ultimately loss of the ROI during the lift-out stage. This problem was overcome by a novel use of the gas deposition capabilities of the FIB. Carbon was deposited to create a supporting pillar structure between the underlying foil surface and the edge of the section. The

micromanipulator needle was then Pt "welded" to the section, and then the FIB was used to remove the material holding the section to the crater wall (Fig. 3c). Finally, the point of contact between the section and the support pillar was ablated away using the FIB. The section was then successfully moved over to a Cu TEM half-grid within the chamber. Once attached to the grid, the section was further FIB-thinned using a 300 pA beam current, reduced to 100 pA for the final thinning to an electron transparent thickness of ~80 nm (Fig 3d).

Serial Sectioning of Residue Material

The initial SEM-EDS study of the crater preserved on foil C2086W,1 identified abundant residue material, including a fragment approximately 20 μm across (Fig. 4a). Isotopic imaging of the fragment using the Washington University NanoSIMS identified a presolar grain (McKeegan et al., 2006) approximately 250 nm in diameter (Fig. 4a). The location of the residue fragment on the interior side-wall of the crater lip meant that it was not possible to prepare a FIB section using the methodology described above for through-cut of an overturned crater lip. For example, the combination of complex sample topography and instrument chamber geometry prohibited deposition of a protective carbon layer over the ROI prior to use of the ion beam. As a result great caution had to be taken when using the FIB to image and mill the material surrounding the residue fragment. The sample topography effectively prevented extraction of a limited ROI containing the presolar grain. Instead, the entire fragment was removed from the interior wall of the crater. The FIB was initially used to make a series of trench cuts around the residue fragment at beam currents between 1000 and 7000 pA (Fig. 4b). A Pt "weld" was

deposited between the fragment and crater wall to reduce the risk of loss during attachment of the micromanipulator needle-point. Once the micromanipulator was attached to the fragment, the Pt "weld" was ablated away using the FIB and the fragment was lifted away (Fig. 4c). The fragment was moved over to an aluminum grid mounted onto a standard pin-mount, where it was attached using Pt "welds" (Fig. 4d). This new location allowed access to the ROI. Protective C and Pt layers were then deposited over the ROI containing the presolar grain. As with the crater lip lift-out, the orientation of the fragment now removed the need for broad trenching to accommodate undercuts using the FIB. Therefore, there was no substantial loss of the surrounding material since it was possible to simply make side-wall FIB cuts and then extract the section with the micromanipulator. Rather than extract only the section containing the presolar grain, multiple sections in series from the fragment were recovered using this side-wall or so-called "bread slicing" technique (Fig. 4e). The sections were then attached to a Cu TEM half-grid and thinned to electron transparency using the FIB conditions described above (Fig. 4f). Using this novel slicing technique, it was possible to extract seven sections from the region of the fragment containing the presolar grain and its surrounding material.

One of the significant disadvantages of applying ultramicrotomy to astromaterials sample preparation is that the diverse range, size and properties of the mineralogical phases within a sample can often result in "plucking" or drop-out of material during the serial slicing, as well as structural deformation within the section. This loss of material can negatively impact the overall mineralogical interpretation of the sample under the TEM. With the FIB serial section technique, the problems are greatly reduced as the sections have a greater structural integrity. However unlike ultramicrotomy, material

between each FIB slice is ablated away resulting in a potential loss of mineralogical continuity when analyzing each section. Despite this disadvantage, serial sectioning using the FIB is a particularly important evolution of the technique that combines site-specific recovery with the benefits (traditionally provided by ultramicrotomy) of serial sections from a single small sample.

Summary

FIB microscopy provides a means to perform integrated studies on individual IDPs and Stardust cometary debris, helping us to explore the relationships between the isotopic, mineralogical and elemental properties at the nano-scale. Such studies are providing new insights into early solar system processes and the interstellar environment (e.g., Floss et al., 2004; Stroud et al., 2004; Bradley et al., 2005; Floss et al., 2006).

As a routine sample preparation technique for meteoritic materials, the "classic" FIB-TEM lift-out has only had limited use outside of these few integrated studies (e.g., Heaney et al., 2001; Lee et al., 2003). This is, in part, due to the reputation of FIB preparation as destructive because the trenching process requires substantial loss of material from the sample. Furthermore, the number of sections that have been harvested from small sample volumes has typically been limited to one or two, comparing most unfavorably with the multiple sections that can be produced by ultramicrotomy. However, the novel "bread-slicing" FIB technique described in this paper for generating serial slices highlights a new capability for extracting multiple sections from a 20 μm diameter residue fragment. This technique is now being applied to IDPs as a method of serial sectioning like ultramicrotomy but with the advantage of less loss of petrographic

information due to grain pluck-out and shattering. FIB microscopy is a mature, yet still evolving, sample preparation technique that is meeting the needs of the next generation of analytical instruments (e.g., the NanoSIMS and SuperSTEM) and samples (e.g., Stardust).

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References

Anderson R. and Klepeis. 2005. Practical aspects of FIB TEM specimen preparation. In *Introduction to focused ion beams – instrumentation, theory, techniques and practice*, edited by Giannuzzi L.A., and Stevie F.A. New York: Springer. pp.173-200.

Bradley J.P. 2005. Interplanetary dust particles. In *Treatise on Geochemistry*, edited by Davis A.M., Holland H.D. and Turekian K.K. Amsterdam: Elsevier. pp. 689-711.

Bradley J.P. 1988. Analysis of chondritic interplanetary dust thin-sections. *Geochimica et Cosmochimica Acta* 52: 889-900.

Bradley J.P. and Brownlee D.E. 1985. Cometary particles: Thin sectioning and electron beam analysis. *Science* 231: 1542-1544.

Bradley J., Dai Z.R., Erni R., Browning N., Graham G., Weber P., Smith J., Hutcheon I., Ishii H., Bajt S., Floss C., Stadermann F., and Sandford S. 2005. An astronomical 2175 Å feature in interplanetary dust particles. *Science* 307: 244-247.

Bradley J.P., Keller L., Thomas K.L., Vander Wood T.B. and Brownlee D.E. 1993. Carbon analysis of IDPs sectioned in sulfur and supported on beryllium films. Proceedings, 24th Lunar and Planetary Science Conference. pp173-174.

Brownlee D.E., Tsou P., Aleon J., Alexander C.M. O'D., Araki T., Bajt S., et al. 2006. Comet 81P/Wild 2 under a microscope. *Science* 314: 1711-1716.

Floss C., Stadermann F.J., Bradley J.P., Dai Z.R., Bajt S., Graham G. and Lea A.S. 2006. Identification of isotopically primitive interplanetary dust particles: A NanoSIMS isotopic imaging study. *Geochimica et Cosmochimica Acta* 70: 2371-2399.

Floss C., Stadermann F.J., Bradley J., Dai Z.R., Bajt S., and Graham G. 2004. Carbon and nitrogen isotopic anomalies in an anhydrous interplanetary dust particle. *Science* 303: 1355-1358.

Giannuzzi L.A. 2006. Reducing FIB damage using low energy ions. *Microscopy and Microanalysis* 12 (S1): 322-323.

Graham G.A., Teslich N., Dai Z.R., Bradley J.P., Kearsley A.T. and Hörz F.P. 2006. Focused ion beam recovery of hypervelocity impact residues in experimental craters on metallic foils. *Meteoritics and Planetary Science* 41: 159-165.

Heaney P.J., Vicenzi E.P., Giannuzzi L.A. and Livi K.J.T. 2001. Focused ion beam milling: A method of site-specific sample extraction for microanalysis of Earth and planetary materials. *American Mineralogist* 86: 1094-1099.

Heymann J.A.W., Hayles M., Gestmann I., Giannuzzi L.A., Lich B. and Subramaniam S. 2006. Site-specific 3D imaging of cells and tissues with a dual beam microscope. *Journal of Structural Biology* 155: 63-73.

Hörz F.P., Bastien, R., Borg J., Bradley J.P., Bridges J.C., Brownlee D.E., Burchell M.J., et al. 2006. Impact features on Stardust: Implications for Comet Wild 2 dust. *Science* 314: 1716-1719.

Ishii H.A. and Bradley J.P. 2006. Macroscopic subdivision of silica aerogel collectors for sample return missions. *Meteoritics and Planetary Science* 41: 233-236.

Ishii H.A., Graham G.A., Kearsley A.T., Grant P.G., Snead C.J. and Bradley J.P. 2005. Rapid extraction of dust impact tracks from silica aerogel by ultrasonic micro-blades. *Meteoritics and Planetary Science* 40: 1741-1747.

Joswiak D.J. and Brownlee D.E. 2006. Non-GEMS silicate glasses in chondritic porous interplanetary dust particles (abstract #2190). 37th Lunar and Planetary Science Conference. CD-ROM.

Keller L.P., Messenger S., Flynn G.J., Clemett S., Wirick S. and Jacobsen C. 2004. The nature of molecular cloud material in interplanetary dust. *Geochimica et Cosmochimica Acta* 68: 2577-2587.

Lee M.R. and Smith C.L. 2006. Scanning transmission electron microscopy using a SEM: Applications to mineralogy and petrology. *Mineralogical Magazine* 70: 561-572.

Lee M.R., Bland P.A. and Graham G.A. 2003. Preparation of TEM samples by focused ion beam (FIB) techniques: application to the study of clays and phyllosilicates in meteorites. *Mineralogical Magazine* 67: 581-592.

Leroux H., Borg J., Troadec D., Djouadi Z. and Hörz F.P. 2006. Microstructural study of micron-sized craters simulating Stardust impacts in aluminium 1100 targets. *Meteoritics and Planetary Science* 41: 181-196.

Matrajt G. and Brownlee D.E. 2006. Acrylic embedding of Stardust particles encased in aerogel. *Meteoritics and Planetary Science* 41: 1715-1720.

McKeegan K.D. Aleon J., Bradley J., Brownlee D., Busemann H., Butterworth A., Chaussidon M., et al. 2006. Isotopic compositions of cometary matter returned by Stardust. *Science* 314: 1724-1728.

Smith J.B., Dai Z.R., Weber P.K., Graham G.A., Hutcheon I.D., Bajt S., Ishii H. and Bradley J.P. 2005. Nitrogen isotopic anomalies in a hydrous interplanetary dust particle (abstract #1003). 36th Lunar and Planetary Science Conference. CD-ROM.

Stadermann et al. 2007. Stardust in STARDUST - the C, N, and O isotopic compositions of Wild 2 cometary matter in Al foil impacts. To be submitted to *Meteoritics and Planetary Science.*

Stevie F.A., Griffis D.P. and Russell P.E. 2005. Focused ion beam gases for deposition and enhanced etch. In *Introduction to focused ion beams – instrumentation, theory, techniques and practice*, edited by Giannuzzi L.A., and Stevie F.A. New York: Springer. pp 53-72.

Stroud R.M. and Zega T.J. 2006. In situ lift-out for coordinated structure-isotope studies. *Scanning* 28: 67.

Stroud R.M., Nittler L.R. and Alexander C.M.O.D. 2004. Polymorphism in presolar Al_2O_3 grains from asymptotic giant branch stars. *Science* 205: 1455-1457.

Teetsov A. and Bradley J. 1986. Micromanipulation of extraterrestrial particles. Proceedings, 17th Lunar and Planetary Science Conference. pp. 883-884.

Warren J.L. and Zolensky M.E. 1994. Collection and curation of interplanetary dust particles recovered from the stratosphere by NASA. In *Analysis of Interplanetary Dust: American Institute of Physics Conference Proceeding 310*, edited by Zolensky M.E., Wilson T.L., Rietmeijer F.J.M. and Flynn G., New York, AIP Press. pp. 245-253.

Westphal A.J., Snead C., Butterworth A., Graham G.A., Bradley J.P., Bajt S., Grant P.G., Bench G., Brennan S. and Pianetta P. 2004. Aerogel keystones: Extraction of complete hypervelocity impact events from aerogel collector. *Meteoritics and Planetary Science* 39: 1375-1386.

Young R.J. and Moore M.V. 2005. Dual-Beam (FIB-SEM) systems. In *Introduction to focused ion beams – instrumentation, theory, techniques and practice*, edited by Giannuzzi L.A., and Stevie F.A. New York: Springer. pp.247-268.

Zega T.J. and Stroud R.M. 2006. In situ lift-out with a focused ion beam/scanning electron microscope: A new technique for creating transmission electron microscope samples of natural and synthetic materials. *Scanning* 28:67-69.

Figure Captions

Fig. 1. (a) 5 kV secondary electron image of IDP L2047,D23 pressed into a high purity gold substrate. The white circle denotes the location of an isotopically anomalous grain that was identified during NanoSIMS isotopic imaging. (b) 5 kV secondary electron image of the IDP after the FIB has been used to make trenches on either side of the region containing the anomalous grain. The Pt "strap" was deposited prior to the initial FIB trenching to reduce the effects of ion beam damage on the surface. The image also shows the side-wall and under-cut FIB cuts made to enable the extraction of the approximately 1 μm thick section. (c) 30 kV ion-induced secondary electron image of the in-situ extraction of the section from the IDP. The needle-point is attached to the section using Pt to form a "weld". (d) 30 kV ion-induced secondary electron image of the section attached to a half-cut Cu TEM grid, again the section is attached to the grid using gas-deposited Pt to form a "weld". (e) 30 kV SEM-STEM bright field image of the section after it has been thinned to electron transparency (approximately 100 nm thick). (f) The transmission mode infrared spectrum acquired from the section identifying a feature around 9 μm that corresponds to layer silicates (marked by the black arrow).

Fig. 2. (a) A 300 kV TEM bright field image of the amorphous carbon grain (white arrow) within the FIB section. The C-rich grain is surrounded by serpentine (serp) and was tentatively identified as the location containing the ^{15}N isotopic enrichment. The grain was confirmed as the carrier of the N isotope anomaly by overlaying the false color NanoSIMS isotope ratio images for ^{12}C (b) and ^{14}N (c) with the TEM image (a). The NanoSIMS images have a point to point resolution of ~50 nm and a pixel dimension of

100 nm^2; the images use an 16-bit color scheme with white-to-yellow representing high intensity and blue-to-black representing low intensity. Additional information concerning the NanoSIMS data is contained in Bradley et al. (2005).

Fig. 3. (a) Composite x-ray elemental maps for Al (blue), Mg (red) and Si (green) overlaid on a 20 kV back-scattered electron image of an impact crater preserved on Stardust Al foil C2091N,1. The cometary residue is denoted by the yellow patches. The lower right edge of the crater lip in the image was selected as a site for FIB extraction of Mg-rich residue. (b) 5 kV secondary electron image of the crater lip containing the region of interest (ROI) after the protective C and Pt straps have been deposited. The arrows point to the bottom of the two side-wall cuts where they milled into the underlying foil. The resulting ROI section is approximately 1 μm thick. (c) 5 kV secondary electron image of the section supported by the gas-deposited C support pillar (false-colored brown) prior to extraction by the in-situ micromanipulator. (d) 5 kV secondary electron image of the section after it has been mounted on the Cu grid and thinned to electron transparency (~80 nm).

Fig. 4. (a) 5 kV secondary electron image of a large micrometer-sized impact penetration preserved on the surface of Stardust Al foil C2086W,1. Insert (i) shows a 5 kV high magnification secondary electron image of the cometary residue fragment that was analyzed using the NanoSIMS at Washington University, St Louis (McKeegan et al., 2006; Stadermann et al., 2007). Insert (ii) shows the 5 kV secondary electron image of the location within the fragment that contains an isotopically anomalous grain (denoted

by the white circle). (b) 5 kV secondary electron image of the residue fragment after the FIB had made the side-walls and undercut to enable the extraction. (c) 5 kV secondary electron image of the fragment as it was extracted from the interior wall of the crater using the in-situ micromanipulator. (d) 5 kV secondary electron image of the fragment after it was re-located onto an aluminum substrate. (e) 5 kV secondary electron image (upper micrograph) and 30 kV ion-induced secondary electron image (lower micrograph) of the extractions of sections 5 and 7 from the fragment using the “bread-slicing” or serial sectioning technique. (f) 5 kV secondary electron image of the Cu grid with three sections extracted from the bulk residue fragments, mounted and further FIB thinned to electron transparency.

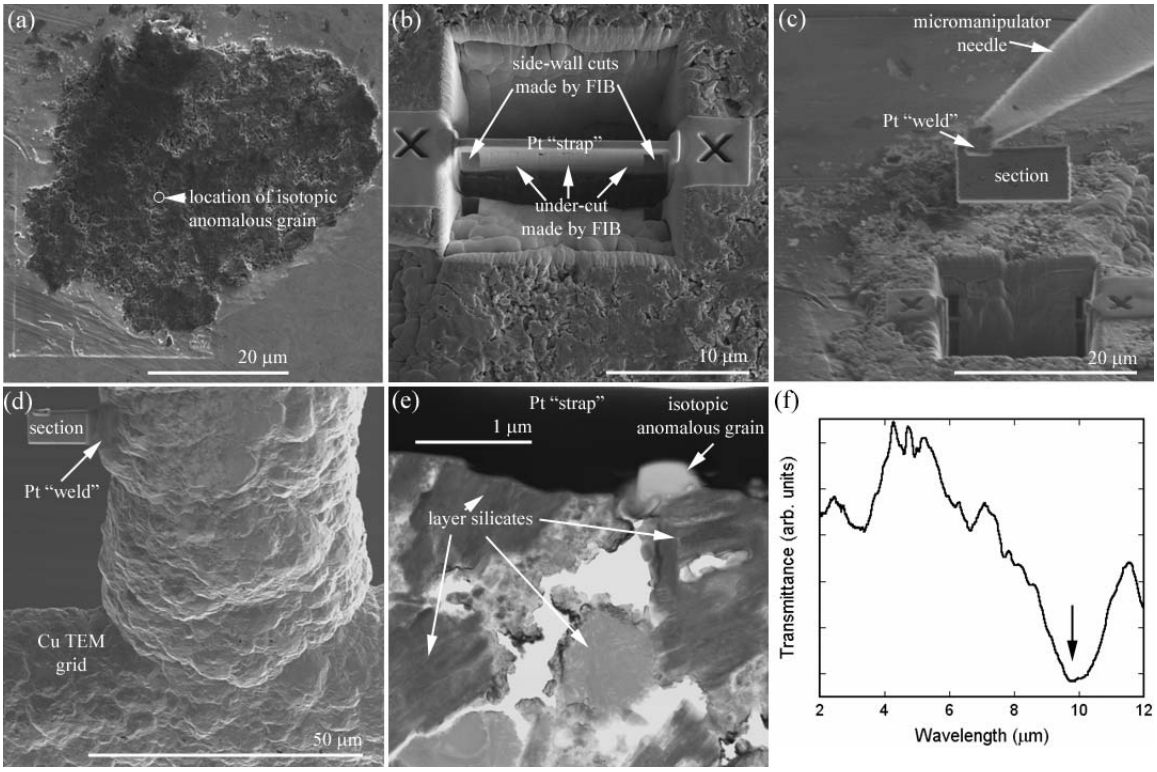
(a)
location of isotopic anomalous grain
20 μm
(b)
side-wall cuts made by FIB
Pt "strap"
under-cut made by FIB
10 μm
(c)
micromanipulator needle
Pt "weld"
section
20 μm
(d)
section
Pt "weld"
Cu TEM grid
50 μm
(e)
Pt "strap"
1 μm
isotopic anomalous grain
layer silicates
(f)
Transmittance (arb. units)
2
4
6
8
10
12
Wavelength (μm)

Fig. 1.

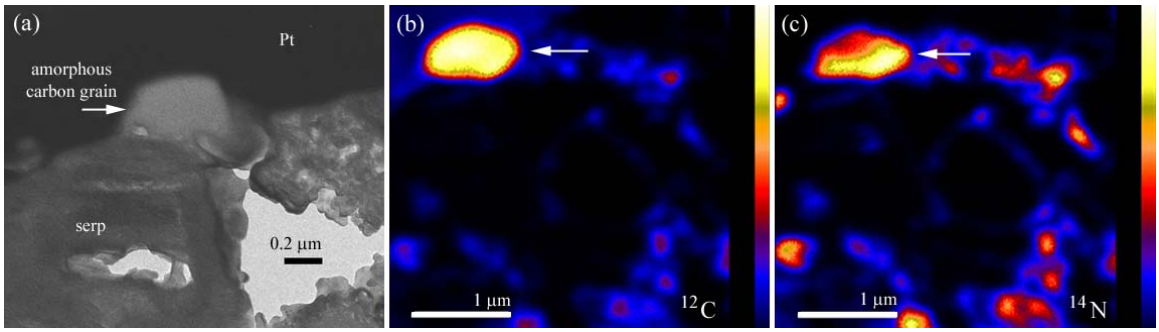
(a)
Pt
amorphous carbon grain
serp
0.2 μm
(b)
1 μm
12C
(c)
1 μm
14N

Fig. 2.

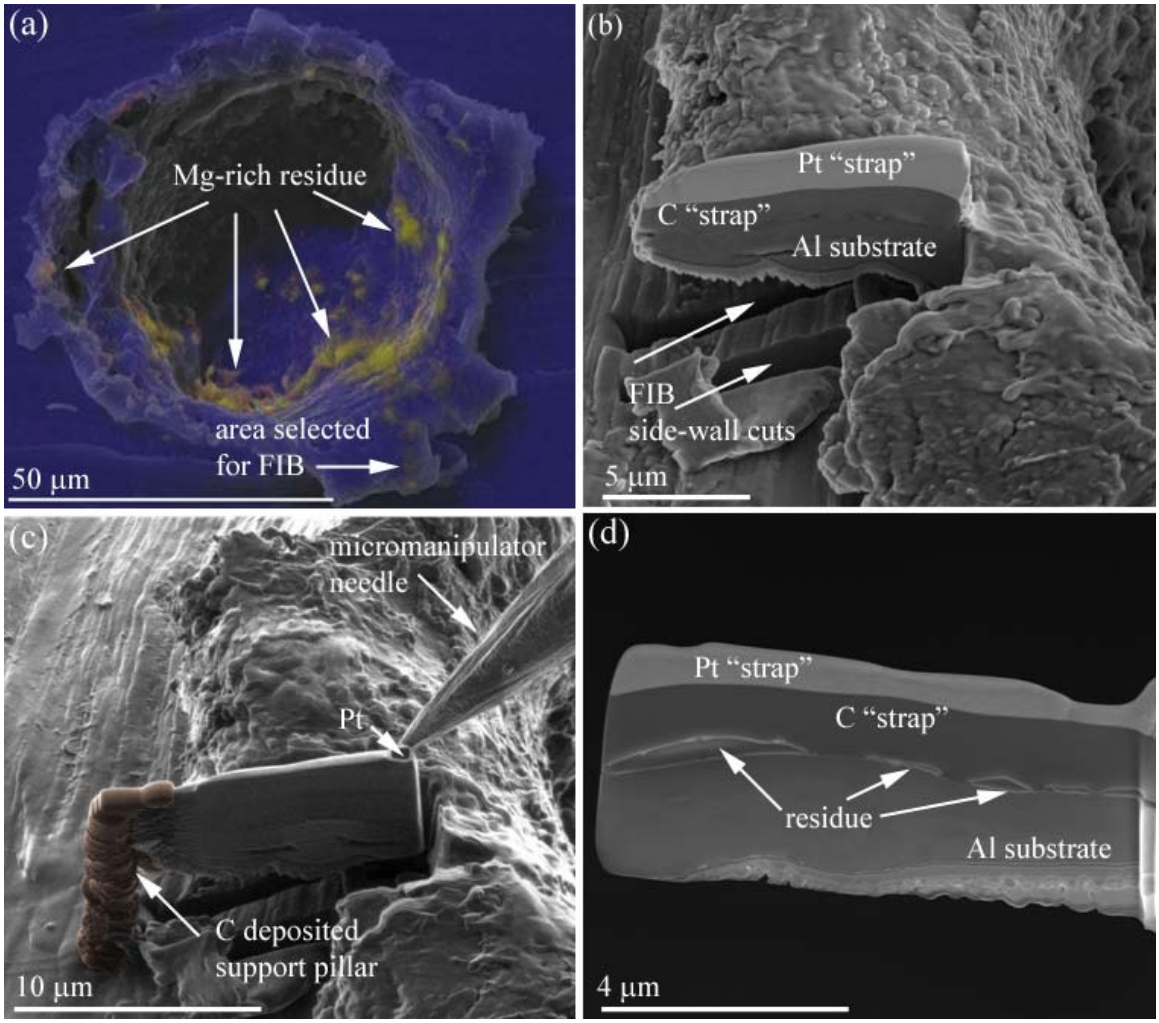
(a)
Mg-rich residue
area selected
for FIB
50 μm
(b)
Pt "strap"
C "strap"
Al substrate
FIB
side-wall cuts
5 μm
(c)
micromanipulator
needle
Pt
C deposited
support pillar
10 μm
(d)
Pt "strap"
C "strap"
residue
Al substrate
4 μm

Fig. 3.

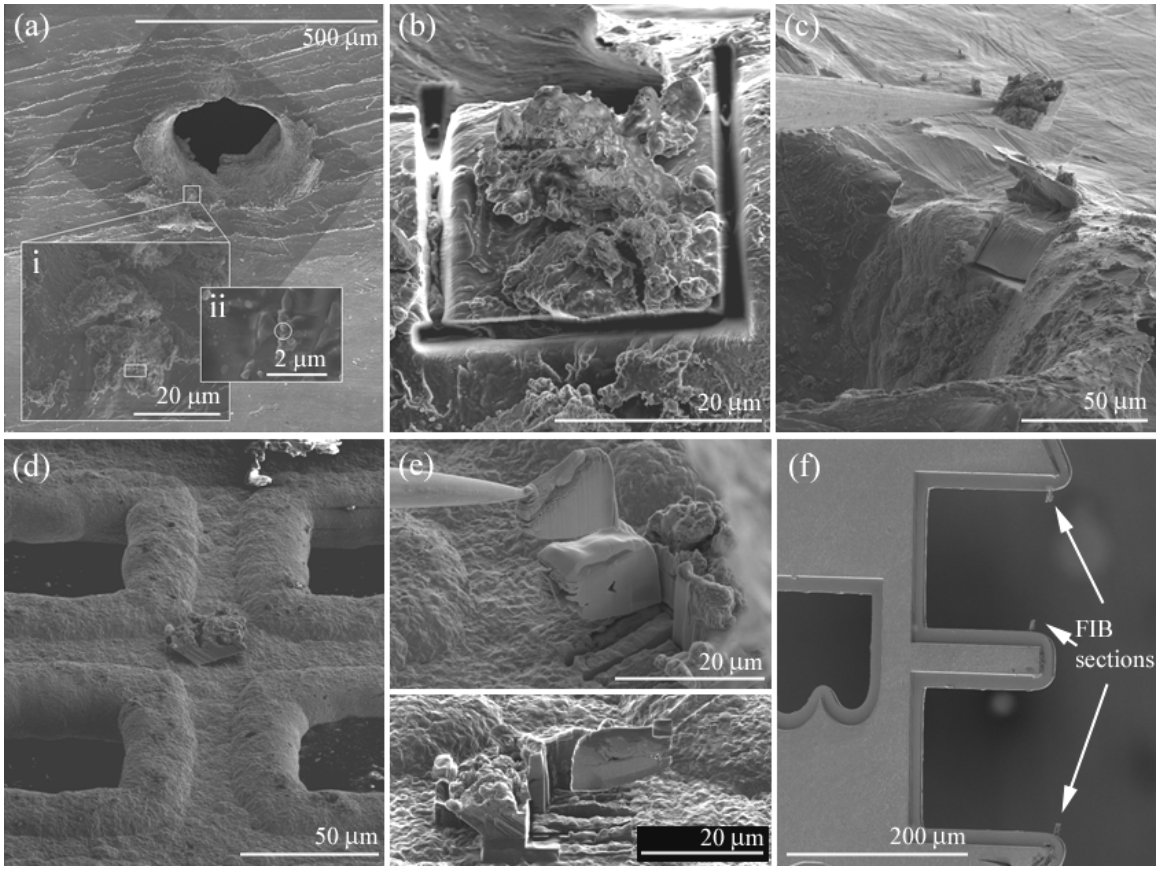
(a)
500 µm
i
ii
2 µm
20 µm
(b)
20 µm
(c)
50 µm
(d)
50 µm
(e)
20 µm
20 µm
(f)
FIB
sections
200 µm

Fig. 4.